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Source apportionment analysis of airborne VOCs using positive matrix factorization in industrial and urban areas in Thailand

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ABSTRACT

Airborne volatile organic compounds (VOCs) concentration data from monitoring stations in industrial and urban (Bangkok) area of Thailand were analyzed using Positive Matrix Factorization (PMF) to identify and elaborate on their source profiles. Analyzed data were obtained from those measured from January 2009 to December 2013 in the study area. The PMF was performed to estimate the contribution of specific source types to ambient concentrations. The most suitable number of factors for a dataset in this study was found to be eleven factors for compositions and characteristics of VOCs. Emission sources of VOCs in industrial area were classified into three to five groups based on their contribution. In industrial area, 42 to 57% of total VOC concentrations were contributed from mobile sources. Contribution to total VOCs concentration from industrial processes and household chemical usage were about 15 to 44% and 3 to 10%, respectively. Moreover, some species of VOCs particularly the ozone depleting substances such as Freon 11, Freon 114 and carbon tetrachloride were found as background concentration in ambient air. As for Bangkok, it was found that most of total VOC concentrations were contributed from mobile source emissions.

Keywords: *Volatile organic compounds (VOCs), positive matrix factorization (PMF), source apportionment, Maptaphut*

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1. Introduction

Volatile organic compounds (VOCs) are important precursors to ozone (Chameides et al., 1992; Carter, 1994). They can form secondary organic aerosols (Ng et al., 2007), and are among the most important ambient carcinogens (McCarthy et al., 2009; U.S. EPA, 2011). Understanding the temporal and spatial characteristics of VOCs gives insight into emission sources to which mitigation measures can be applied to improve air quality and reduce human health impacts. Hazardous air pollutants (HAPs) are generally defined as pollutants that are known or suspected to cause cancer or other serious health effects, or to cause harm to the environment (OAQPS, 1998). The 1990 Clean Air Act Amendments (CAAA), Section 112, seek to reduce human exposure to HAPs by defining a statutory list of these compounds (U.S. EPA, 1997). More than 80% of the compounds on the federal HAPs list are toxic volatile organic compounds (VOCs). Some examples of sources of personal exposure to toxic VOCs include household cleaners, vehicle exhaust, gasoline vapors, dry–cleaned clothes, and environmental tobacco smoke (ETS). The majority of toxic VOCs in ambient air originate from sources that emit to the outdoors, such as drycleaners, petrochemical complexes, power plants, and vehicle emissions (U.S. EPA, 2000; Liu et al., 2008). Solvent usage and vehicular emissions (including gasoline, diesel vehicular emissions and gasoline evaporation) are two major contributors to local VOCs (Ling et al., 2011).

Maptaphut complex consists of five industrial estates and a port. The complex is located in Maptaphut sub–district, Rayong province, East of Thailand. Maptaphut Industrial Estate (MTPIE) is the biggest industrial estate, which is the most important manufacturing base for petrochemicals, chemicals, steel, metals and refineries of the country. It was developed in 1989 by the state enterprise, Industrial Estate Authority of Thailand (IEAT), Ministry of Industry. Rapid development of industry cause many environmental and health problems such as lack of water and air quality effect. Ambient air monitoring operated by Pollution Control Department (PCD) routinely report concentration of more than forty VOCs in ambient air of Maptaphut. Monitoring results from 2007–2012 indicated that there were five VOC compounds namely benzene, 1,3–butadiene, chloroform, dichloromethane and 1,2–dichloroethane which their annual concentrations were higher than the Thailand's ambient air quality standards (PCD, 2013).

Receptor models provide a theoretical and mathematical framework for quantifying source contributions. One of the receptor oriented model used to identify contribution of emission sources of air pollutants is Positive Matrix Factorization (PMF). This technique is used to derive a set of source composition profiles, each identifying a mix of compounds associated with a particular category of emissions (e.g., on–road emissions, solvent manufacturing, etc.). PMF analysis could be applied to small data sets to enable source location and apportionment for air pollutants in small scale investigations of localized pollution problems (Chan et al., 2011). Algorithms used in the PMF model have been peer reviewed by leading air and water quality management scientists (U.S. EPA, 2008).

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There are several other receptor models besides PMF. The most commonly used ones are UNMIX, PCA/APCS, and CMB (Leuchner and Rappengluck, 2010). Each approach has its advantages and limitations as described in several comparison studies (e.g. Willis, 2000; Anderson et al., 2002; Miller et al., 2002; Song et al., 2008; Viana et al., 2008a). In general, the PMF method showed good agreement with the UNMIX model (e.g. Anderson et al., 2002; Jorquera and Rappengluck, 2004) and performed very well in comparison to CMB and PCA/APCS. In contrast to UNMIX, PMF, in particular the flexible multi–linear engine, can solve multi–linear problems with the possibility of implementing many kinds of constraints, e.g. individual data points can be weighted (Willis, 2000; Viana et al., 2008b), despite the increasing risk of putting additional distance between statistical model and physical reality (Willis, 2000). Miller et al. (2002) found out that in comparison to the other three modeling approaches, the extracted factors from the PMF analysis represented the major sources that were used to generate the simulated data most closely. The lack of the non–negativity constraint is another significant limitation of PCA/APCS and CMB (Anderson et al., 2002).

This study is aimed to identify source of VOCs in Maptaphut sub–district, Rayong province, Thailand. Ambient VOC concentration data from Pollution Control Department (PCD) were analyzed by the receptor modeling using the Positive Matrix Factorization (PMF). Monitored data from January 2009 to December 2013 (5 years) were analyzed. This long–term investigation assists in increasing number of data for PMF evaluation.

2. Material and Methods

2.1. Information about the monitoring sites

Dindaeng is a roadside monitoring station, located in Bangkok. This site is located in the central business district (CBD) of the metropolis. Traffic volume of Dindaeng road was about 98 000 vehicles/day. Therefore, it is appropriate to be used as background to represent a profile of VOCs influenced by vehicle emissions. Spatial location of VOC monitoring site at Dindaeng was as shown in Figure 1.

There were seven VOCs monitoring stations in Maptaphut area. Monitoring stations were located at Health Promotion Hospital Maptaphut (HMTP), Ban Ta Kuan Public Health Center (BTKH), Wat Nong Fap School (WNFS), Muang Mai Maptaphut (MMTP), Map Chalut Temple (MCLT), Ban Plong Community (BPLC) and Nop Pakate Village (NPKV), respectively. Since VOC ambient standards in Thailand were set up with their concerns on direct health impact, monitoring sites were selected following locations of the community areas. The nearest communities to industrial complex were given priority due to the fact that VOCs were mainly emitted from non–stack sources. Therefore, high concentrations of VOCs could occur in the vicinity of their emission sources. This assumption was used as the worst case scenario in analysis of VOCs impact in the study area. Furthermore, selected monitoring stations were located covering all directions from industrial complex. Characteristics and spatial distribution of VOCS monitoring sites were as presented in Table 1 and Figure 1, respectively.

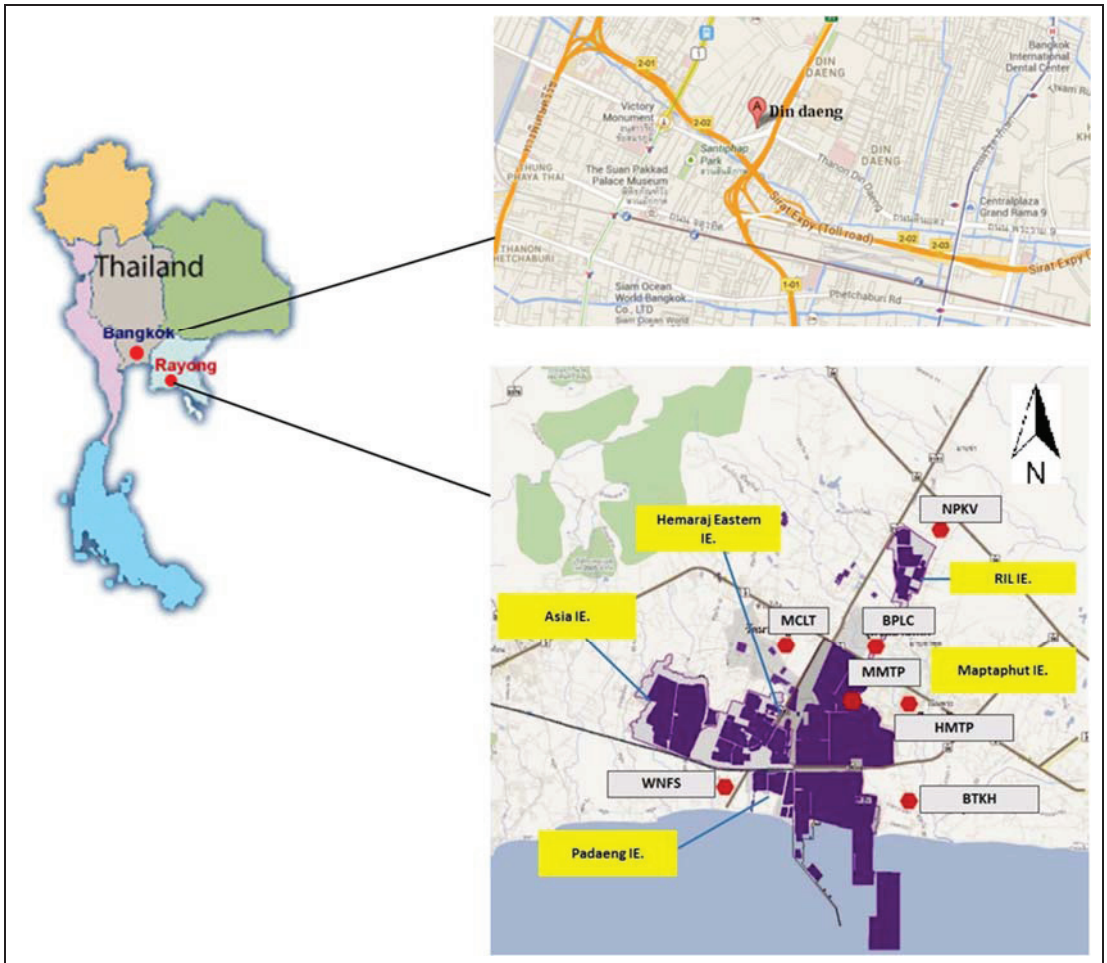

Figure 1. *Location of the study area and sampling sites (IE=Industrial estate).*

Table 1. *Characteristics of VOCs monitoring sites in Maptaphut*

Monitoring Sites	Direction from Maptaphut Complex	Distance from Maptaphut Complex (km)	Distance to the Nearest Road (km)	Distance to the Main Road (km)
Health Promotion Hospital Maptaphut (HMTP)	Northeast	0.62	0.03	0.64 (NO. 3)
Ban Ta Kuan Public Health Center (BTKH)	Southeast	0.98	0.01	3.23 (NO. 3)
Wat Nong Fap School (WNFS)	Northeast	1.50	0.02	0.02 (NO. 3392)
Muang Mai Maptaphut (MMTP)	Northeast	0.32	0.08	0.29 (NO. 3)
Map Chalut Temple (MCLT)	Northeast	1.68	0.04	1.55 (NO. 3)
Ban Plong Community (BPLC)	Northeast	0.08	0.05	0.63 (NO. 3)
Nop Pakate Vilage (NPKV)	Northeast	0.22 [a]	0.01	0.62 (NO. 36)

[a] *The distance of Nop Pakate Village station was measured from RIL industrial complex*
Traffic volume of the main roads: Route No. 3=36 095 vehicles per day, Route No. 36=39 143 vehicles per day, Route No. 3392=20 160 vehicles per day

This industrial area is normally influenced by two major prevailing wind directions. The southwest wind direction is dominant wind during wet season (April to October). Almost all of the monitoring stations were situated downwind from the industrial complex during this period except the WNFS station which was located in the southwestern part of the study area. Opposite wind direction is blown from northeast direction during dry season (November to March). However, during the monitored one–year period of 2013, prevailing winds were approximately 70% from the southwest and 30% from the northeast. Wind rose diagram of the year 2013 is presented in Figure 2 as an example. During this time, monitoring stations are located upwind position from the industrial complex. However, it was found that there were no temporal differences in measured total VOCs concentrations. Example of monthly variations of total VOCs concentrations at MMTP station was as shown in Figure 3. This finding could be explained by the fact that the monitoring sites were also located at the downwind position from RIL petrochemical estate during the dry season (Figure 1).

2.2. Data collection

Ambient VOC concentration data were measured by Pollution Control Department (PCD). Data used in this study were measured in Maptaphut sub–district, Rayong province and Bangkok province, Thailand from January 2009 to December 2013. VOC samples were collected by 6 liter evacuated canisters (at 0.05 mmHg) and were analyzed using gas chromatography/mass spectrophotometry (GC/MS).

The analytical method was based on US. EPA TO15 (U.S. EPA, 1999). When the canisters were opened to the atmosphere, the VOC sample was introduced into the canisters by the differential pressure between atmospheric pressure and vacuum pressure inside each canister. With a flow controller, the sub–atmospheric sampling system maintained a constant flow rate from full vacuum to within about 713.80 kg/m^2 or less below ambient pressure. Canister flow rate was controlled by flow controller and was adjusted to 5.5×10^{-8} m^3/s for 24–h sampling. After collecting the ambient VOCs, the sample canister was pressurized by humidified nitrogen about 14.2x10^3 kg/m^2 in order to prevent the contamination entering the sample canister. Samples were transferred to the thermal desorption unit, working as a pre–concentrator prior to being sent to GC/MS (Thepanondh et al., 2011).

2.3. PMF model description

Positive Matrix Factorization (PMF) is a multivariate factor analysis tool that decomposes a matrix of speciated sample data into two matrices factor contributions and factor profiles. Identification of emission source contributions can be performed using measured source profile information, wind direction analysis, and emission inventories. The method is reviewed briefly here and described in greater detail elsewhere (Paatero and Tapper, 1994; Paatero, 1997; Jeong et al., 2011).

PMF version 3.0 analyses the correlation between measured concentrations of chemical species, assuming that highly correlated compounds come from the same source. The PMF can be explained as the linear product of a source matrix and a contributing matrix. The two matrices are obtained by an interactive minimization algorithm: PMF involves constrained maximization of a weighted object function. PMF model is expressed as follows:

$$X_{ij} = \sum_{k=1}^{p} g_{ik} f_{kj} + e_{ij} \quad (1)$$

An ambient data set can be viewed as a data matrix X of i by j dimension, in which i is the number of samples, j is the chemical species that were measured, p is the number of factors that best characterize the VOC composition at a site, g_{ik} is the contribution of each source factor to each sample, f_{kj} is the chemical composition profile of each source factor, and e_{ij} is the residuals. The number of factors, p, in the real dataset is generally unknown. Choosing the best modeled number of factors for a dataset is the most critical decision to the interpretation of the PMF results (Ulbrich et al., 2009). There are several qualitative metrics for making the determination of the number of factors. In this study, the criterion of a solution with the least rotation (lowest maximum value of Rotmat) was applied. The best solution which provided the least rotation of dataset used in this study was calculated as 11.

Results are constrained by a penalty function so that no sample can have a negative source contribution and no species can have a negative concentration in any source profile. Strength of PMF is that it can individually weight each data point. This feature allows adjusting of the influence of each data point, depending on the confidence in the measurement. By individually weighing data, samples with some species missing or below the detection limit, do not need to be excluded as a whole. The uncertainty can be adjusted to minimize effect of missing or below detection limit species on final solution (Paatero and Tapper, 1994; Paatero, 1997).

The PMF solution minimizes the object function *Q*, based on these uncertainties (*u*):

$$Q = \sum_{i=1}^{n}\sum_{j=1}^{m}\left[\frac{X_{ij} - \sum_{k=1}^{p} g_{ik}.f_{kj}}{u_{ij}}\right]^2 \quad (2)$$

In this study, the U.S. EPA equation approach was used to estimate the uncertainty associated with each data point and missing concentration values were replaced by average concentration of the species (U.S. EPA, 2008). As for each compound, if more than 25% of number of samples were reported as missing value or lower than method detection limit, the compound was excluded from PMF analysis.

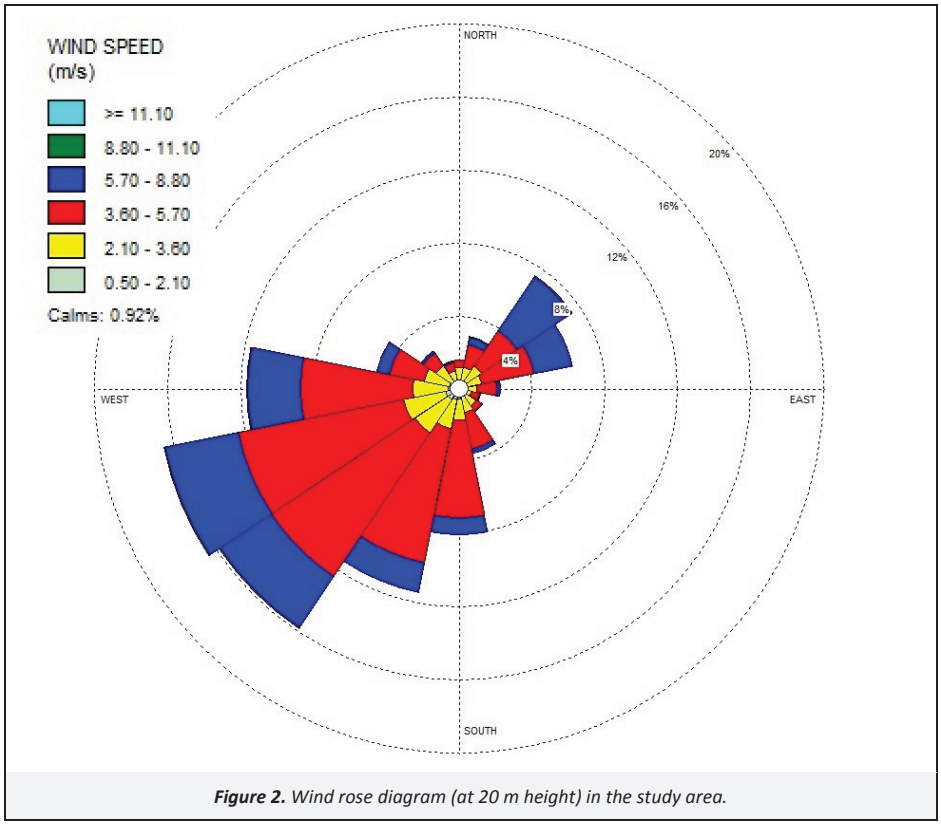

Figure 2. *Wind rose diagram (at 20 m height) in the study area.*

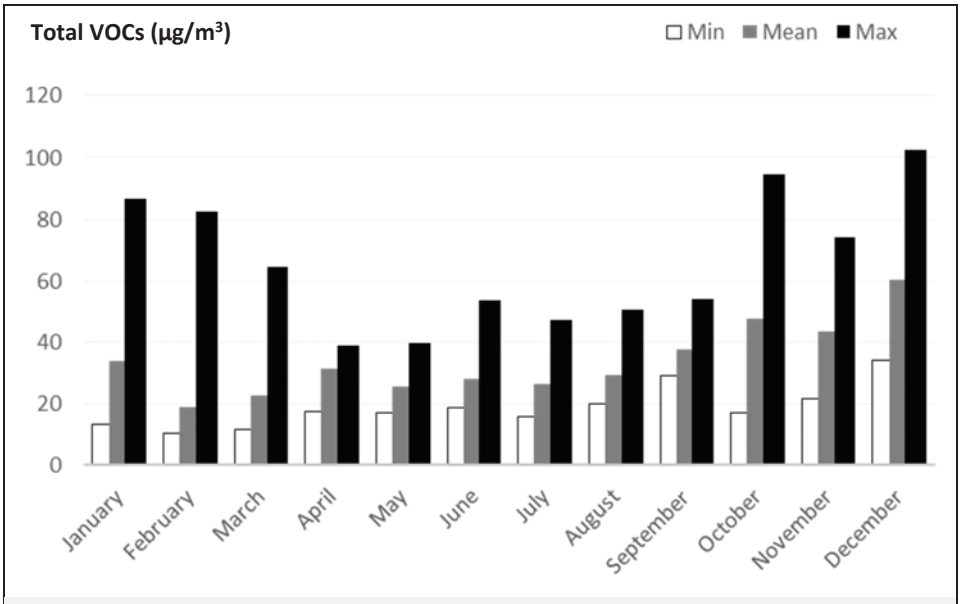

Figure 3*. Monthly variation of total VOC concentrations in Maptaphut industrial area (data from 2009 to 2013).*

3. Results

3.1. PMF results at Maptaphut

Results of PMF analysis from eleven factors were grouped according to presence of signature compounds of each emission category. Possible emission sources of each factor were determined by using relationship among VOCs and their percentage of contribution in the same group. Emission sources consisted of factor taking into consideration that benzene, toluene and total xylenes were signature of mobile sources. Vinyl chloride and 1,2–dichloroethane which are used as raw materials in the PVC (poly vinyl chloride) industry were used as signature of this industrial group. Industrial process emission were dominated by toluene, styrene and 1,3–butadiene were signature of this source group. Chemical use in household source emissions were dominated by 1,2–dichloropropene, 1,1–dichloroethylene, acrylonitrile, 1,2–dibromoethane, 1,1,1–trichloroethane and benzyl chloride. Background concentrations in ambient air source emissions were dominated by Freon 11, Freon 12, Freon 113, Freon 114 and carbon tetrachloride (Kim et al., 2005; Liu et al., 2008; Ling et al., 2011).

Results from PMF analysis at each receptor site were presented in Figures S1 to Figure S8 (see the Supporting Material, SM). It should be noted that percent contributions for each compound were used to indicate contributions of each emission source to individual VOC concentrations. For example, in Figure S1, percent contribution of vinyl chloride and 1,2–dichloroethane from industrial process (PVC) group were analyzed as about 80% and 75%, respectively. However, only the percentages of contribution to total VOC concentrations from each emission source were recalculated in this study.

Analytical results for each receptor site were as follows: At HMTP, possible emission sources could be grouped into five factors as shown in Figure S1 (see the SM). Factor 1 was identified as mobile source emissions and was estimated that its contribution to total VOC concentrations was approximately 44.3%. Factor 2 was identified as industrial process (PVC) emission. Contribution of this source group to total VOC concentrations was estimated as 4.9%. Factor 3 represented industrial process emissions with its contribution of about 33.2% to total VOCs. Factor 4 was classified as chemical use in households. Contribution of this group to total VOCs was about 2.6%. Factor 5 was background concentrations in ambient air. It was estimated that about 10.5% of total VOC concentrations appeared in this group. Unidentified was about 4.5% of total VOCs. At MMTP, possible emission sources could be grouped into five factors as shown in Figure S2 (see the SM). Each factor was represented similar groups with those at HMTP monitoring site. Percentage of their contribution to total VOCs concentration were 42.8%, 8.1%, 28.3%, 9.8% and 5.9% for Factor 1 to Factor 5, respectively. Unidentified was about 5.1% of total VOCs. At NPKV possible emission sources could be grouped into five factors as shown in Figure S3 (see the SM). Each factor was represented by similar groups with those at HMTP and MMTP monitoring sites. Percentage of their contribution to total VOC concentrations were 48.1%, 6.6%, 23.8%, 8.7% and 5.1% for Factor 1 to Factor 5, respectively. Unidentified was about 7.5% of total VOCs. At BTKH, possible emission sources could be grouped into five factors as shown in Figure S4 (see the SM). Each factor represented by a similar group with those previous monitoring sites. Percentage of their contribution to total VOCs concentration were 45.7%, 11.5%, 25.3%, 4.7% and 8.7% for Factor 1 to Factor 5, respectively. Unidentified was about 4.1% of total VOCs. At BPLC, possible emission sources could be grouped into three factors as shown in Figure S5 (see the SM). Factor 1 represented the mobile source emissions. Contribution of this group to total VOC concentrations was about 42.4%. Factor 2 was identified as industrial process (PVC) emission. Its contribution to total VOC concentrations was estimated as 7.2%. Factor 3 was grouped as industrial process emissions. About 45.0% of total VOCs were estimated to be related to this emission source. Unidentified was about 5.3% of total VOCs. At MCLT, possible emission sources could be grouped into four factors as shown in Figure S6 (see the SM). Factor 1 was identified as mobile source emissions. Its contribution to total VOC concentrations was about 56.4%. Factor 2 was classified as industrial process emissions. About 15.7% of total VOCs were estimated to be contributed by this source. Factor 3 represented chemical use in households. It was estimated that about 6.9% of total VOC concentrations originated from this source. Factor 5 represented the chemicals found in background ambient air. Contribution of this group was estimated as 7.4% to total VOC concentrations. Unidentified was about 13.5% of total VOCs. At WNFC, possible emission sources could be grouped into four factors as shown in Figure S7 (see the SM). Factor 1 was identified as mobile source emissions. About 57.6% of total VOCs concentration was estimated to be emitted from this source. Factor 2 was identified as industrial process (PVC) emissions. Contribution of this source to total VOCs concentrations was estimated as 5.9%. Factor 3 represented industrial process emissions. It was estimated that about 13.1% of total VOC concentrations was originated from this group. Factor 4 was chemical use in households. Its contribution to total VOC concentrations was estimated at about 6.2%. Unidentified was about 17.2% of total VOCs.

Generally, results of emission sources from PMF analysis at Maptaphut indicated that about 42.4–57.6% of total VOC concentrations were contributed by mobile source emissions. This finding could be explained by the fact that most of the monitoring sites were located close to the roads to serve accessibility of the monitoring network. This finding could be used to evaluate representativeness of monitoring sites in this industrial area. Results from this study revealed that these existing monitoring sites may not be appropriate to use in assessing the impact from emissions of industrial sources.

3.2. PMF results at Dindaeng

The results of Positive Matrix Factorization (PMF) analysis for Dindaeng (Bangkok) area could be grouped into four factors as shown in Figure S8 (see the SM). Possible emission sources of each factor were determined by using the relationship among VOCs and their percent contributions within the same group. At Dindaeng, Factor 1 was identified as gasoline vehicle exhaust emissions which was mainly attributed to benzene, total xylenes and ethyl benzene. About 19.4% of total VOC concentrations was estimated to be originated from this source. Factor 2 was identified as diesel vehicle exhaust. Its contribution to total VOC concentrations was about 13.7%. Contribution of other mobile sources of emission, i.e., evaporation was 17.4% to total VOCs. Factor 3 was classified as chemical use in households. About 31.5% of total VOCs were estimated to be contributed from this group. Factor 4 was represented by chemicals found in background ambient air. Examples of chemicals in this group were Freon 11, Freon 12, Freon 113, Freon 114, and carbon tetrachloride. Contribution of this group to total VOC concentrations was estimated at 7.6%. Unidentified was 10.3%. Contributions of each emission source to total VOC concentrations at both areas in this study were summarized in Table 2.

Similar with Maptaphut industrial area, percent contribution to total VOC concentrations at Dindaeng from mobile sources was about 50.5%. About 31.5% of total VOCs at Dindaeng was contributed from chemical use in households category. This result indicated the ability of PMF analysis in this study in distinguishing major emission groups in each area. High contribution of household source to total VOCs concentration emphasizes the need for further study to analyze the behavior and pattern of chemical usage in Bangkok.

Table 2. *Summary of percentage of emission source contribution to total VOCs concentration*

Site	Mobile Source	Industrial Process Source [a]	Chemical Use in Household Source	Background Concentration in Ambient Air	Unidentified
Maptaphut Station					
HMTP	44.3%	33.2% (4.9% PVC)	2.6%	10.5%	4.5%
MMTP	42.8%	28.3% (8.1% PVC)	9.8%	5.9%	5.1%
NPKV	48.1%	23.8% (6.6% PVC)	8.7%	5.1%	7.5%
BTKH	45.7%	25.3% (11.5% PVC)	4.7%	8.7%	4.1%
BPLC	42.4%	45.0% (7.2% PVC)			5.3%
MCLT	56.4%	15.7%	6.9%	7.4%	13.5%
WNFS	57.6%	13.1% (5.9% PVC)	6.2%		17.2%
Dindaeng	50.5%		31.5%	7.6%	10.3%

[a] *Percentage of PVC industry contributions are shown in parenthesis*

In order to serve a Thai's regulation for VOCs management, we further examined contribution of each emission source to individual VOC compounds using the same dataset. Benzene was selected as an example of air toxic compound for analysis of its source profile and contribution in Maptaphut and Dindaeng. It was found that mobile source emissions contributed to benzene's concentration of about 0.5 to 3.7 and 1.3 to 7.7 μg/m^3 at Maptaphut and Dindaeng, respectively. These contributions were exceed the Thai ambient air quality standard for benzene (1.7 μg/m^3) at both locations. Therefore, efforts to control not only industrial sources but also mobile emission sources are necessary even in the industrial area.

4. Conclusions

Positive Matrix Factorization (PMF) model was applied to identify possible emission sources and to evaluate contribution of each emission source to total VOC concentrations. Comprehensive VOC monitoring data, measured in Maptaphut (industrial area) and Dindaeng (urban area) from January 2009 to December 2013 were analyzed in this study. Analytical results revealed that total VOC concentrations in industrial area were contributed by both mobile and industrial emission sources. As for urban site, results indicated that mobile sources comprised the largest source of total VOCs in the urban area (about 51% of total VOC concentrations). The other principle emission source was chemicals use in households (about 31% of total VOC concentrations). Generally, results from PMF analysis at Maptaphut indicated that about 42.4–57.6% of total VOC concentrations were contributed by mobile source emissions. Therefore, it was estimated that about 43.37 μg/m^3 from total amount of 85.88 μg/m^3 of total VOCs concentration in Bangkok and about 10.34–40.83 μg/m^3 from total amount of 24.39–70.88 μg/m^3 of total VOC concentrations in Maptaphut area originated from mobile source emissions. This profile supported our hypothesis that existing VOC monitoring stations in Maptahut industrial area did not fully represent the impact from industrial emissions. As for health impacts, Maptaphut has been declared a pollution control zone by the Thai government which means pollution emissions must be restricted. Results from environmental monitoring are used to evaluate the outcomes of mitigation measures in controlling emissions from industrial sources. Problems with the representativeness of monitoring stations which are discovered in this analysis should be considered in order to have better planning management of air pollution in this industrial area. Contribution of emissions from PVC factory to total VOC concentrations in this study area was also much interested due to carcinogenic property of vinyl chloride and 1,2 dichloroethane which are used as raw materials for PVC manufacturing. Taking into consideration that monitoring sites in this study were located in the community area, a further study of VOCs exposure assessment should be conducted to reveal health impacts of air toxics in this industrial area.

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Supporting Material Available

Source profiles (percentage of factor total) resolved from PMF in Health Promotion Hospital Maptaphut (HMTP) monitoring station (Figure S1), in Muang Mai Maptaphut (MMTP) monitoring station (Figure S2), in Nop Pakate Village (NPKV) monitoring station (Figure S3), in Ban Ta Kuan Public Health Center (BTKH) monitoring station (Figure S4), in Ban Plong Community (BPLC) monitoring station (Figure S5), in Map Chalut Temple (MCLT) monitoring station (Figure S6), in Wat Nong Fab School (WNFS) monitoring station (Figure S7), in Dindaeng (Bangkok) monitoring station (Figure S8). This information is available free of charge via the internet at http://www.atmospolres.com.

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